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Transient Techniques for Battery Impedance Measurements

Small-Amplitude Exponential Perturbation Technique

Prepared by

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1 July 1981

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AIR FORCE SYSTEMS COMMAND
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This technical report has been reviewed and is approved for publication. Publication of this report does not constitute Air Force approval of the report's findings or conclusions. It is published only for the exchange and stimulation of ideas.

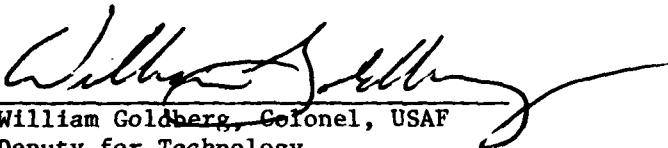
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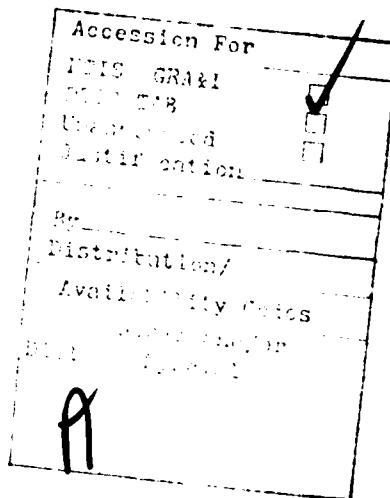
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20. ABSTRACT (Continue on reverse side if necessary and identify by block number) A perturbation technique is reported for measuring the impedance of battery cells under conditions of controlled potential. The small amplitude exponential perturbation (SAEP) technique is applicable over an extremely wide frequency range and appears to be the method of choice for measuring the impedance of battery cells that contain very little stored electrochemical energy.		

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I. INTRODUCTION

The proper operation of battery cells invariably depends on a number of internal physical and chemical reactions occurring at rates that are sufficient to sustain cell performance. These reactions typically involve charge transfer processes at the electrodes, as well as diffusional transport of materials to the active electrode surfaces. Kinetic measurements permit determination of the relative importance of these processes in controlling cell performance. The most general method for making these kinetic measurements is to measure the electrical impedance of the battery cell as a function of frequency. The rates of the various processes that affect the cell voltage are inferred directly from the frequency dispersion of the cell impedance.

A number of techniques have been used to measure the impedance of battery cells. The most commonly used is that of applying a sinusoidally varying ac signal to the battery cell and monitoring the cell response in terms of amplitude and ac phase shift. This ac method is relatively easy to use, but if data are required over a wide frequency range or at very low frequencies, it becomes somewhat cumbersome. Other techniques for impedance measurements of battery cells incorporate perturbing functions other than sinusoidal ac. For example, in the galvanostatic transient technique¹ a step change in the current passing through the cell is applied, and the response of the cell to the current change is measured. The relationship between the change in cell current and the voltage response gives the cell impedance. This technique is particularly useful when the cell contains appreciable stored capacity, since in this case controlling cell current is much easier than controlling cell voltage. However, when the cell contains very little stored capacity, any measurement attempted under conditions of constant current may change the cell voltage by a large amount and thereby appreciably alter the chemical state of the cell. In this situation, it is desirable to employ a potentiostatic

¹A. H. Zimmerman and M. R. Martinelli, Transient Techniques for Low Frequency Impedance Measurements, TR-0079(4970-10)-1, The Aerospace Corporation, El Segundo, Calif (6 October 1978).

technique that involves the application of a controlled perturbation to the cell potential.

We have developed and applied such a technique to battery cells. This technique is called small amplitude exponential perturbation (SAEP) and involves perturbing the cell voltage with a small amplitude (<5 mV) exponential signal while measuring the current response of the cell. Again, the cell impedance is obtained from the relationship between voltage and current. This technique can be used to measure the impedance of battery cells at any voltage or state of charge that is accessible to them, although very large currents (and power supplies) may be involved when the cell has appreciable active electrochemical capacity.

II. THEORY OF SAEP

Any potentiostatic transient technique for measuring impedance employs a transient potential function $V(t)$. This potential function is applied as a perturbation to a battery cell that has the initial potential V_0 . The cell current is initially $I_0 + I_N(t)$, where I_0 is the steady-state current at V_0 , and $I_N(t)$ is any change in current resulting from depletion of the stored electrochemical capacity of the cell at the initial voltage. After the perturbation $V(t)$ is applied, the current is $I_0 + I_N(t) + I(t)$. For this analysis to be correct, the amplitude of $V(t)$ must be sufficiently small that $I_N(t)$ does not change appreciably in response to $V(t)$. This means that typically $V(t)$ should be less than 5 mV in amplitude. In addition, the time constant associated with $I_N(t)$ must be much greater than that associated with $I(t)$ so that they can be separated in time.

The impedance as a function of time is then directly given by Ohm's law

$$Z(t) = \frac{V(t)}{I(t)} \quad (1)$$

However, the cell impedance is more conveniently analyzed in the frequency domain. Laplace transformation of $V(t)$ and $I(t)$ permits us to obtain the impedance as a function of frequency.

$$Z(\omega) = \frac{V(\omega)}{I(\omega)} \quad (2)$$

where $V(\omega)$ and $I(\omega)$ are the Laplace transforms of voltage and current, respectively.

The digital Laplace transforms required are calculated from the voltage and current data,

$$F(\omega) = \int_0^\infty f(t) \exp(-j\omega t) dt = \sum_i \int_{t_i}^{t_{i+1}} f_i(t) \exp(-j\omega t) dt \quad (3)$$

which are digitized by computer into arrays having i data points, each corresponding to a given time. The function $f_i(t)$ fits the data for $f(t)$ in the interval t_i to t_{i+1} and may be any convenient function that fits the data. Functions used for $f(t)$ include linear, quadratic, and exponential forms as follows.

1. Linear: $f_i(t) = A_i t + B_i$

$$A_i = \frac{f(t_i) - B_i}{t_i}$$

$$B_i = \frac{[f(t_{i+1}) - t_{i+1}/t_i f(t_i)]}{1 - \frac{t_{i+1}}{t_i}} \quad (4)$$

2. Quadratic: $f_i(t) = L_i t^2 + M_i t + N_i$

$$M_i = [\frac{\Delta f_{12}}{\Delta t_{12}^2} (\frac{t_1^2}{t_{i+2}} - t_{i+2}) - \frac{\Delta f_{13}}{t_{i+2}}] [1 - (t_{i+2}) \frac{\Delta t_{12}}{\Delta t_{12}^2} + \frac{t_i}{t_{i+2}} (\frac{t_i \Delta t_{12}}{\Delta t_{12}^2} - 1)]^{-1}$$

$$L_i = \frac{\Delta f_{12}}{\Delta t_{12}^2} - M_i (\frac{\Delta t_{12}}{\Delta t_{12}^2}) \quad (5)$$

$$N_i = f(t_i) - L_i t_i^2 - M_i t_i$$

where

$$\Delta t_{12} = t_i - t_{i+1}$$

$$\Delta t_{12}^2 = t_i^2 - t_{i+1}^2$$

$$\Delta f_{12} = f(t_i) - f(t_{i+1})$$

$$\Delta f_{13} = f(t_i) - f(t_{i+2})$$

3. Exponential:

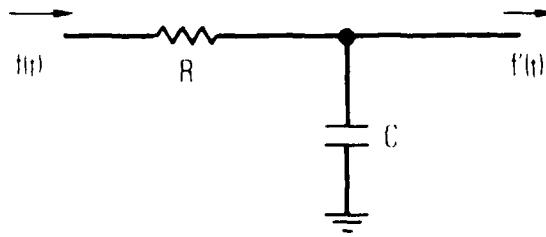
$$f_i(t) = C_i \exp(-a_i t) \quad (6)$$

$$a_i = \ln \left[\frac{f(t_{i+1})/f(t_i)}{t_i - t_{i+1}} \right]$$

$$C_i = f(t_i) \exp(a_i t_i)$$

The exponential function of Eq. (6) gave the best results for all data that were not near a point where $f(t)$ crossed zero. A listing of a Fortran program for doing the transformations that give the impedance is provided in the Appendix.

Actual experimental data contain noise; in particular, 60-Hz noise may pose a problem when small changes in voltage or current are being monitored. The simplest way to eliminate this kind of noise is with an RC filter.



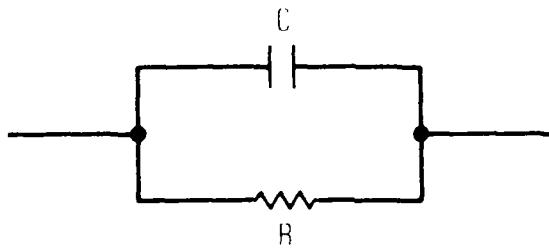
However, the transfer function of this filter must then be deconvoluted from the data. This is easily done in the frequency domain simply by multiplying the transformed function by the inverse filter function transform

$$f(\omega) = f'(\omega) (1 + j\omega\tau_F) \quad (7)$$

where $\tau_F = RC$ is the filter time constant.

III. RESULTS

The impedance of battery cells below 1 kHz is generally capacitive in nature, behaving as an equivalent parallel RC circuit where the values of R and C may have a complicated dependence on frequency. The results of an SAEP experiment on a simple dummy cell consisting of the RC circuit



where $C = 1 \text{ F}$ and $R = 10 \Omega$ are examined first. For simplicity, let us assume that $V_0 + V_N(t)$, the initial cell voltage, is zero. An increasing exponential perturbation having amplitude α and time constant τ is applied to the cell

$$V(t) = \alpha(1 - e^{-t/\tau}) \quad (8)$$

$V(t)$ and the current response of the dummy cell $I(t)$ are indicated in Fig. 1 for $\tau = 2\text{s}$, $R = 10 \Omega$, and $C = 1 \text{ F}$. $I(t)$ is given by the relationship

$$I(t) = \frac{\alpha}{R} \left[1 - \left(1 - \frac{RC}{\tau}\right) e^{-t/\tau} \right] \quad (9)$$

Note that the values of R and C do not influence the time constant for current decay, but only control the amplitude of the current transient. From the time-dependent voltage and current functions, the impedance is calculated, with the results shown in Fig. 2 in the complex plane. The results in Fig. 2 agree with the theoretical result for the impedance

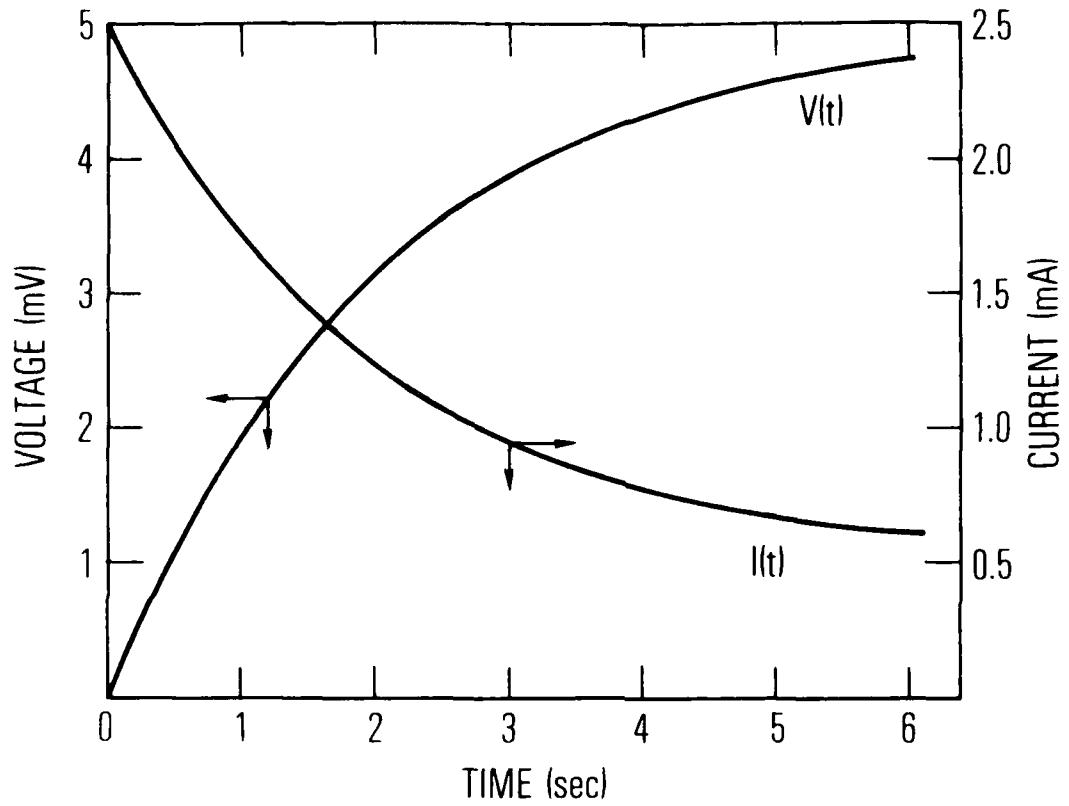


Fig. 1. Voltage Perturbation and Response Current for Dummy Cell

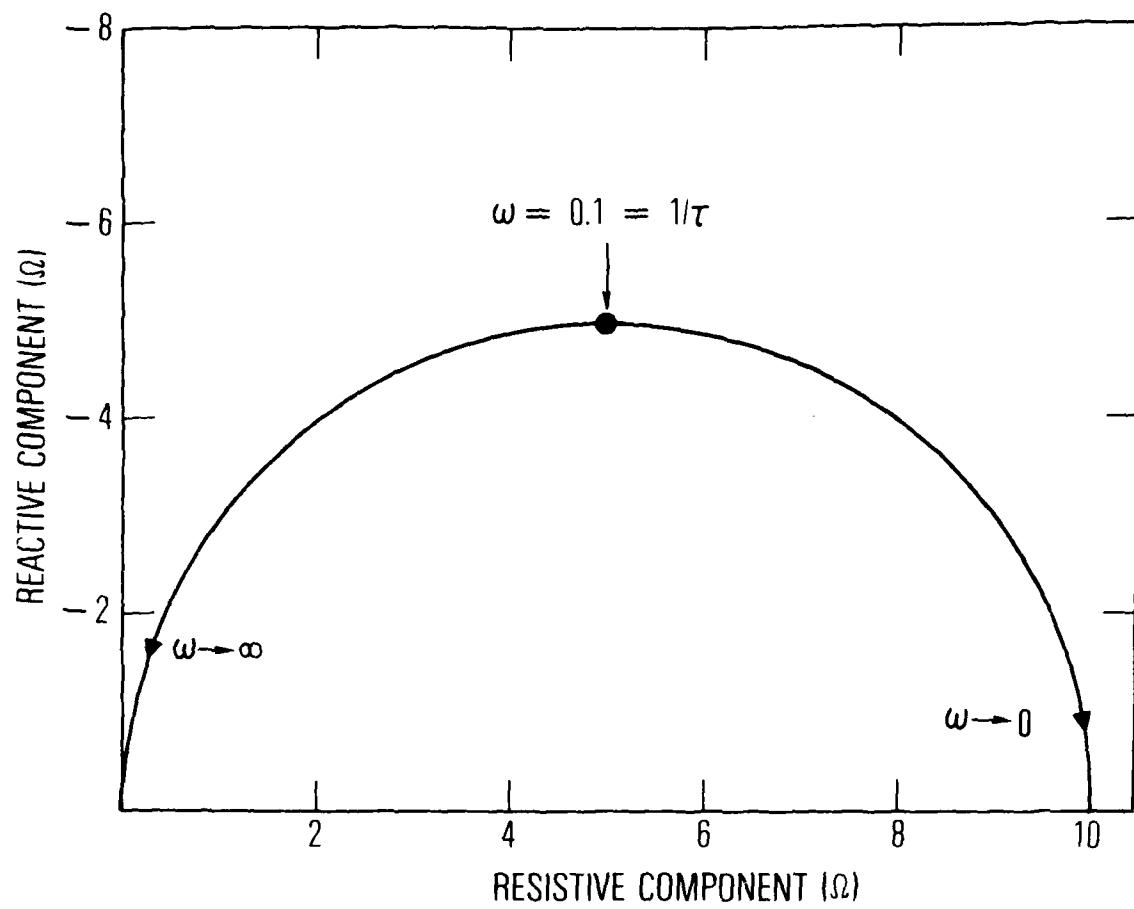


Fig. 2. Impedance of Dummy Cell from Data of Fig. 1

$$Z(\omega) = \frac{R}{1 + j\omega RC} \quad (10)$$

This simple example illustrates that the expected current response for a battery cell consists of a rapid rise to a maximum, followed by a decay to a steady-state current that is different from the initial current by the amount α/R . The magnitude of the peak current is controlled by the relative time constants of the exponential perturbation and the cell, and is given by $\alpha C/\tau$ in the preceding example. Thus, the experimental maximum transient current can be controlled simply by controlling the perturbation time constant.

The results obtained when an exponential perturbation is applied to a nickel cadmium cell are shown in Fig. 3. The nickel cadmium cell used was a 10-Ah prismatic cell, and the initial cell voltage was 0.5 V. The impedance is indicated in the complex plane in Fig. 4. In making these measurements, it was found that signal to noise became relatively poor unless the time constant of the applied perturbation was the same order of magnitude as the relaxation time for the battery cell. With this general requirement satisfied, the SAEP technique provides a convenient method for making impedance measurements on battery cells over an extremely wide range of frequencies, under conditions of battery operation for which potential control is acceptable.

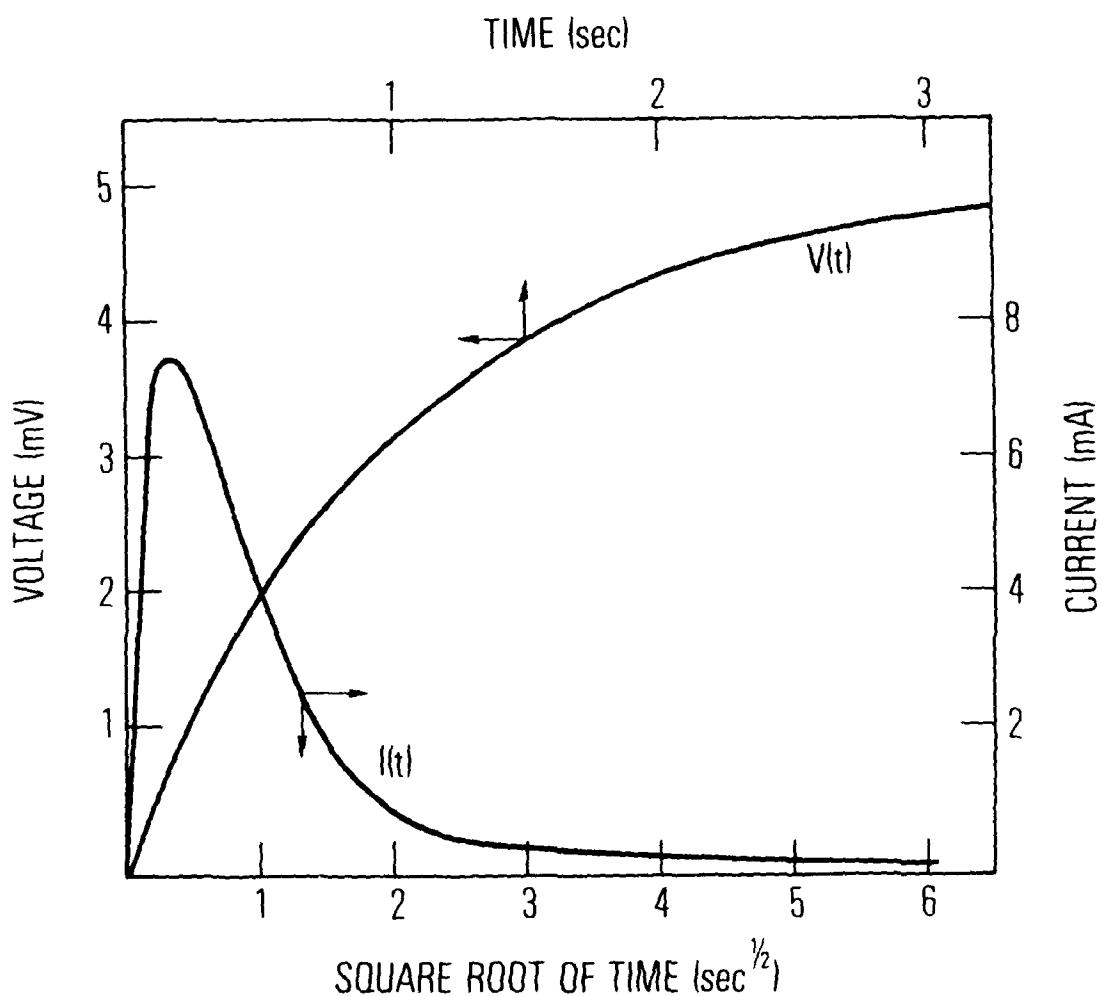


Fig. 3. Voltage Perturbation and Current Response for Nickel Cadmium Cell at 0.5 V

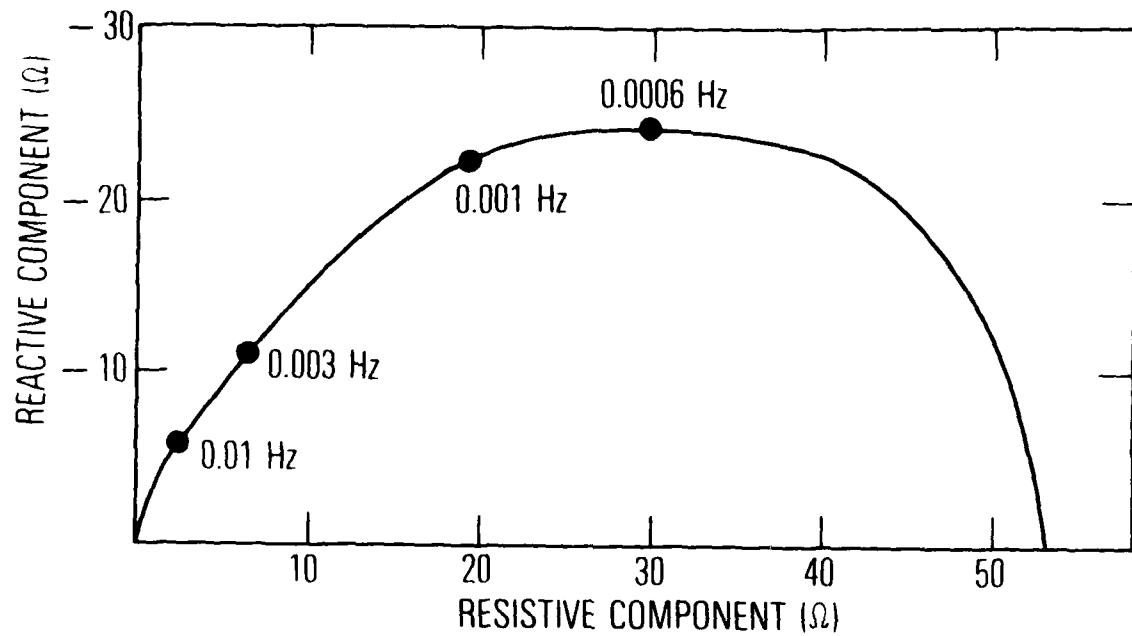


Fig. 4. Impedance of Nickel Cadmium Cell from Data of Fig. 3

IV. CONCLUSIONS

The SAEP technique has been developed and applied to measuring the impedance of battery cells under conditions of controlled potential. This appears to be the optimum method for measuring the impedance of battery cells that contain little stored electrochemical capacity.

APPENDIX

FORTRAN PROGRAM FOR SAEP IMPEDANCE CALCULATION

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PROGRAM SAEP (INPUT, OUTPUT, TAPES=OUTPUT) FREQUENCY FROM THIS PROGRAM CALCULATES IMPEDANCE AS A FUNCTION OF FREQUENCY FROM AN EXPONENTIAL VOLTAGE PERTURBATION AND ITS CURRENT RESPONSE USING THE LAPLACE TRANSFORMATION TECHNIQUE. THE LAPLACE TRANSFORMATION MAY BE INCLUDED IN THE CURRENT RESPONSE, AND IS DECONVOLUTED FROM THE TRANSFORM.

THE PROGRAM FITS THE DATA TO A MAXIMUM OF TWO DIFFUSION PROCESSES. IT REQUIRES INITIAL GUESSES FOR THE FITTING PARAMETERS WHICH ARE TIME CONSTANTS α_1 AND α_2 , WARBURG COEFFICIENTS S_1 AND S_2 , AND CAPACITANCES C_1 AND C_2 AS WELL AS ELECTROLYTE RESISTANCE R_Z .

DATA INPUTS ARE EXPERTMENT IDENTIFICATION NUMBER
 MPROB = NUMBER OF VOLTAGE DATA POINTS
 NDATA = NUMBER OF CURRENT DATA POINTS
 TSAMP = NOMINAL PERTURBATION TIME CONSTANT IN SECONDS
 AMPV = AMPLITUDE OF EXPONENTIALLY CHANGED AT INFINITE TIME IN MA
 FILTTC = TIME CONSTANT OF FILTER USED FOR VOLTAGE/TIME DATA (SEC)
 FILTIV = TIME CONSTANT OF FILTER USED FOR CURRENT/TIME DATA (SEC) AND
 VV(I), TEV(I) ARE ORDERED PAIRS CORRESPONDING TO TIME (SEC) AND
 EXFONENTIAL VOLTAGE DATA (MA) AT ZERO TIME AND SHOULD BE A NOMINALLY INCREASING
 XTE(I), TSI(I) ORDERED PAIRS CONSISTING OF THE SQUARED ROOT OF
 TIME AND CURRENT DATA (MA), AND SHOULD BE ZERO AT
 AND XTC(I) AT INFINITE TIME
 COMMON/A1(5,2),A2(5,2),YD(800),XS(4000),F(200),RZ,
 COMMON/2/NP200,PES(7)
 COMPLEX ZM(2,200),VH,XW,P,XFF,VFF
 EQUIVALENCE (ONS,ZM)
 DATA SWITCH=1 GIVES WINDING2, WFIN/V ONLY
 DATA SWITCH=2 GIVES IMPEDANCE CALCULATION AND FIT TO MODEL
 SWITCH=3 GIVES IMPEDANCE CALCULATION AND FIT TO MODEL
 IF(EOF,100,301)

301
 WRITE(6,60001) MPP OR
 WRITE(6,60001) TSAMP, FILTTC
 WRITE(6,60001) AMPV, AMPI
 WRITE(6,60001) TSV(I), VV(I), I=1,NDATV
 WRITE(6,60001) TSI(I), XI(I), I=1,NDATI
 WRITE(6,60001) TSV(J), VV(J), I=1,NDATV
 WRITE(6,60001) TSI(J), XI(J), TSI(J)
 WRITE(6,60001) AT1
 WRITE(6,60001) AT2
 WRITE(6,60001) AT3
 WRITE(6,60001) AT4

```

206      TSI(J)=TSI(J)*J*J*(1-EX)*(1-EX*(-TSI(J)/TSAEP))
210      XI(J)=XI(J)-AMP1*(1-EX)*(1-EX*(-TSI(J)/TSAEP))
223      HQITE(6,700)
230      N=N+1
233      X(N)=WINI*(N-1)*WINC1
237      NY=N-1
241      X(N)=X(INN)+WINC2
246      CONTINUE
252      H=1.0/(X(N)+*2)
254      F(N)=H*(2*3*141592654)
255      P=CMPPLX(X,W)
256      CALL LPXFRE(MDATA,V,TSV,VY,W,VW)
257      CALL LPXFRE(MDATA,TSI,XI,W,XIW)
258      ADD ZERO FREQUENCY COMPONENT TO TRANSFORM
263      VH=AMPV/P-VW
264      CALL AMP1/(P+(*1+P*TSAEP)) * XIW
265      COMPUTE INVERSE FILTER FUNCTIONS
271      XF=CMPLX(1.0,W*FILT)
272      VFF=CMPLX(1.0,W*FILT)
273      ZM(N)=(VW*VFF)/(XIW*XF)
274      TF((X(N))/GE.*WFIN-WINC2) GO TO 401
275      GO TO 325
280      CONTINUE
285      WRITE(6,6008)
290      WRITE(6,700)
295      WRITE(6,6009)
300      DO 420 I=1,N
305      I=I+1
310      I2=I+1
315      WRITE(6,6010) I,X(I),03S(I1),08S(I2),F(I)
320      CONTINUE
325      C0F(SWITCH,EN,0.) GO TO 300
330      READ(5,202) A1,S1,C1,A2,S2,C2,R2
334      NE2*N
335      CALL GAULL
336      NPROB=1
337      CALL GAUHH
338      GO TO 300
339      CONTINUE
340      STOP
341      FORMAT(3I5,5F10.5)
342      2002 FORMAT(7F10.5)
343      2003 FORMAT(8F10.5)
344      6000 FORMAT(1X,1S,1F10.5)
345      6001 FORMAT(1X,1S,1F10.5)
346      6002 FORMAT(1X,1S,1F10.5)
347      6003 * T=F10.5 *(SECOND)
348      6003 * FORMAT(5X,1VOLTAGEL7X,(CURRENT CHANGE AT LONG T
349      6003 * YES IS 1.0.5)
350      6004 FORMAT(5X,1INPUT DATA VOLTAGE 7X,(SQRT TIME())
351      6005 FORMAT(15X,1INPUT DATA VOLTAGE 7X,(SQRT TIME())
352      6006 FORMAT(5X,1INPUT DATA CURRENT 7X,(SQRT TIME())
353      6007 FORMAT(10X,1INPUT DATA VOLTAGE 7X,(SQRT TIME())

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6008 FORMAT(1X,[IMPEDANCE DATA])
6009 FORMAT(8X,[DATA POINT] 1X,[1/SQRT(OMEGA)] ,7X,[REAL
      *RY Z[8X[FREQUENCY(HZ)] ]
6010 FORMAT(10X,15,4(4X,F14.6))
7000 FORMAT(1X)
ENC

```

PROGRAM LENGTH INCLUDING I/O BUFFERS

EINZELN ASSESSMENTS

STATEMENT	ASSIGNMENTS	301	320	-	-	000231	360	00247
300	-	000364	00033	-	-	000464	20001	00506
401	-	000357	430	-	-	000515	20001	00517
402	-	000351	00035	-	-	000547	60005	00557
6002	-	000524	60003	-	-	000573	60009	00577
6006	-	000561	60007	-	-	000567	60008	00567
6010	-	000611	70006	-	-	000615	70006	00615
BLOCK NAMES AND LENGTHS								
SAEP	-	000745	-	012742	A	-	001446	2
VARIABLE	ASSIGNMENTS							
AMPI	-	000670	AMFV	-	-	000667	A1	000000302
C1	-	000602	C2	-	-	000651	F	0012431504
FILTV	-	000672	T	-	-	000677	I	FILTC
JNDATV	-	000674	MPROB	-	-	000663	INDATI	000000301
P	-	000664	NN	-	-	000600	S01	000000302
S1	-	000647	PEFS	-	-	000675	OBS	000000301
TSV	-	000602	S2	-	-	000612	SWTCH	000000302
W	-	000630	VFF	-	-	000627	TSI	000000301
X1	-	000676	WBRK	-	-	000666	YHINC1	000000301
ZH	-	000652	WINIT	-	-	000661	XFF	000000302
		000650	XIN	-	-	000657	YHINC2	000000301
		000645	XS	-	-	000655	ZHINC1	000000301

STAFF OF CONSTANTS

START OF TEMPORARIES

START OF INDIRECTS

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ପ୍ରକାଶକ ପତ୍ର ପତ୍ର

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SUBROUTINE MODEFL(NPROB,0,YMD(400),XS(4000),FS(2000),
COMMON/A1/S1,C1,A2,S2,C2,V(200),TSV(200),TSI(200))
DIMENSION G(1,1),Z(1,1),ZC1,ZC2,Z1,Z2,G
COMPLEX URA,UI
COMPLEX CTAH
DATA UR,UI/1.,0.,0.,0./
DATA CTAH/1./
DATA ZC1/1./
DATA ZC2/1./
DATA S1/1./
DATA C1/1./
DATA A2/1./
DATA S2/1./
DATA C2/1./
DATA V/1./
DATA TSV/1./
DATA TSI/1./
DATA NG2/2/
DO 120 I=1,NG2
XX=X(I)
ZC1=-UI*XX**2/CI
ZM1=S1*XX*(1-UI)+CTANH(A1+CSQRT(A1+CSQRT(UI)/XX))
Z=ZC1+ZM1/(ZC1+ZM1)
Z2=0.2*E0.0+.1
IF(A2<=0.1*XX**2/C2) GO TO 110
ZM2=ZC2*XX*(1-UI)+CTANH(A2+CSQRT(A2+CSQRT(UI)/XX))
Z=ZC2+ZM2/(ZC2+ZM2)
110 CONTINUE
120 CONTINUE
END

```

210
211 22222222222222222222

6 A2
66666666666666666666
SUBROUTINE PARTIAL(NGD0,NG,NG0)
COMMON N,X(2C),OBS(40),YMD(800),XS(4000),F(200),R2
DIMENSION I(1),NGD0(NG,NG),NO
DO 120 I=1,NO
P=PS12=.5/PEPS(I)
QSAVE=Q(I)
Q(I)=QSAVE+PEPS(I)
CALL MODEL(NPROB,I,NGD0(I,I),NG,NG,NO)
CALL = QSAVE-PEPS(I)
CALL MODEL(NPROB,0,YMD(1),NG,NG)
Q(I)=QSAVE
DO 120 J=1,NG
DO 120 J=1,NG
CONTINUE=(NGD0(J,I)-(NGD0(J,I)-YMD(J)))*PEPS12
120 RETURN
FNC

SUBPROGRAM LENGTH
000105

FUNCTION ASSIGNMENTS

STATEMENT ASSIGNMENTS

BLOCK NAMES AND LENGTHS

VARIABLE	ASSIGNMENTS	-	012742	2	-	000010
O3S	012431S01 I	=	000101	JPEPS12	-	000104
X	000311S01 PEPS	=	0002571S12	YMD	-	000102
	0000311S01 XS	-	002571S12	-	-	001131S11

START OF CONSTANTS

000067
SIAPT OF TEMPORARIES

000071
START OF INDIRECTS

000075
UNUSED COMPILER SPACE
011200

```

COMPLEX FUNCTION CTANH(Z)
COMPLEX UI,7
DATA UI/0.1618/1./60.1/
TF(CABS(Z).GT.1.0)
CTANH=-UI*C SIN(UI*Z)/C OS(UI*Z)
RETURN
TF(CABS(Z).GT.39.1)
CTANH=(1.-CEXP(-Z)).*Z/(1.+CEXP(-Z.*Z))
RETURN
CONTINUE
CTANH=CMPLX(1.,0.)
RETURN
END

```

SUBPROGRAM LENGTH 000143	FUNCTION ASSIGNMENTS STATEMENT ASSIGNMENTS 1 - 003047 2 - 0	BLOCK NAMES AND LENGTHS CTANH - 000143	VARIABLE ASSIGNMENTS UI CTANH - 003137	START OF CONSTANTS 000115	START OF TEMPORARIES 000123	START OF INDIRECTS 000137	UNUSED COMPILER SPACE 011200
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አሁን በዚህ የዕለታዊ ስምምነት እና የሚከተሉት ስምምነቶች ተመርሱ ይችላል

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      SUBROUTINE IXFRM(INDAT,I,VTH,WVH)
C THIS SUBROUTINE COMPUTES THE LAPLACE TRANSFORM AT ANGULAR FREQUENCY
C OF THE SET OF ORDERED PAIRS T AND VT CORRESPONDING TO TIME AND
C FUNCTION VALUE AT THAT TIME. AT INFINITE TIME AND CANNOT CROSS ZERO.
C DIMENSION T(20),VT(20)
C COMPLEX VTH,CA,CI,CIP,CA1,CI1,CIP1
C VTH=CMPLX(C,.C.)
C DO = NDAT-1, 1, -1
      DO = IDO-1, 1, 1
      IP = I+1
      IF(VT(IP).EQ.0.C) VT(IP)=0.00001
      IF(VT(IP).LT.0.C) GO TO 150
      IF(VT(IP).GT.0.C) GO TO 150
      IF(VT(IP).EQ.VT(IP-1)) GO TO 150
      AA=ALOG(VT(IP)/VT(IP-1))/((T(IP)-T(IP-1))
      CA=CMPLX(A,AA)
      CT=CMPLX(0,0)-W*T(IP)
      CTP=CMPLX(0,0)+CEXP(CI*WT(IP))
      VTH=YTH+(VTH*CTP-CT*VTH)/CA
      GO TO 100
  100 IF(T(IP).EQ.0.C) T(IP)=0.00001
      DT=(VT(IP)/T(IP))-DT*VT(IP)/(1-DT)
      B=(VT(IP)-DT*VT(IP))/(1-DT)
      A=(VT(IP)-B)/T(IP)
      CA1=CMPLX(0,0)
      C1=CEXP(-CA1*T(IP))/CA1
      CIP1=CEXP(-CA1*(T(IP)+1.)/CA1)
      VW=VTH*(CIP1*(T(IP)+1./CA1)-CIP1*(T(IP)+1./CA1))+B*(CI1-CIP1)
      CONTINUE
      VW=YTH+VTH*(IP)*CEXP(CIP1)/CA
      RETURN
      END

```

SUBPROGRAM LENGTH						
000420						
FUNCTION ASSIGNMENTS						
STATEMENT ASSIGNMENTS						
100 - 000255	101	-	000312	150	-	000122
BLOCK NAMES AND LENGTHS						
BLPKFRM - 000420						
VARIABLE ASSIGNMENTS						
A - 000415	R	-	000417	CA	-	000404
CI - 000406	CIP	-	000402	CIP1	-	000405
DI - 000416	I	-	000413	IH	-	000414
START OF CONSTANTS						
000315						

